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(54) **Lubricating oil composition comprising a specific base oil and an oxidation inhibitor**

(57) Described is a lubricating oil composition which is excellent in oxidation stability and comprises a base oil and an oxidation inhibitor, wherein the lubricating base oil is a mineral oil-derived lubricating oil having a saturated hydrocarbon content of 80% by mass or more and the viscosity-density constant of the saturated hydrocarbon component is 0.79 or less.

EP 0 896 050 A1

Description

- [0001] The present invention relates to a lubricating oil composition which comprises a specified base oil composition and additive providing a synergistic effect conducive to an highly improved oxidation stability.
- 5 [0002] A good oxidation stability has long been required for a lubricating oil.
- [0003] Recently, a lubricating oil has been inevitably put in use under a sever condition because the lubricating oil has been used in a less amount in a device which tends to be reduced in size and weight, resulting in a higher load being applied to the portions to be lubricated.
- 10 [0004] In particular, a lubricating oil in an internal combustion engine tends to deteriorate due to its reactivity with air or combustion gas because it is exposed to a high temperature regions, for example, between a piston and a cylinder. Further, the recent increase in automobiles, which are provided with a ternary catalyst as a measure to meet the regulation at exhaust gas, compels the internal combustion engine to operate harder. Furthermore, from the standpoint of energy saving, since the body of an automobile is required to have a lighter weight so as to improve the fuel efficiency, the crank case is being downsized and accordingly the amount of crank case oil to be introduced is being reduced. As
- 15 a result, a lubricating oil for the internal combustion engine is inevitably used in a more sever condition than before.
- [0005] In addition, the number of gas engines which use a gas, such as natural gas, LPG or a cracked gas, has been increasing recently, and the temperature at which a lubrication oil is used in these gas engines is becoming even higher.
- [0006] Meanwhile, in the field of lubricating oils for industrial use, oils such as an air compressor oil, a steam turbine oil and a gas turbine oil are required to be maintenance-free for use of long period in a condition leading to a strong
- 20 oxidative deterioration.
- [0007] Accordingly, in recent times there is a strong demand for the development of a lubricating oil having a higher oxidation stability, whereas the prior-art technique, which is based on a combination of a mineral or synthetic base oil and an oxidation inhibitor such as hindered phenol, aromatic amine or zinc dithiophosphate, has proved to be inadequate.
- 25 [0008] For this reason, the present invention was made, and it is hence the object of the present invention to provide a lubricating oil composition having a higher oxidation stability.
- [0009] The present inventors have examined the oxidation stability of a lubricating oil composition containing an oxidation inhibitor to clarify the effect of the base oil composition and the oxidation inhibitor on the oxidation stability. As a result, they have established the present invention based on the discovery that the use of a base oil having a specific
- 30 composition in combination with an oxidation inhibitor brings about a high oxidation stability due to the synergistic effect of the combination.
- [0010] According to the present invention, there is provided a lubricating oil composition comprising a base oil and an oxidation inhibitor, wherein the lubricating base oil is a mineral oil-derived lubricating oil having a saturated hydrocarbon content of 80% by mass or more and the viscosity-density constant of the saturated hydrocarbon component is 0.79 or less.
- 35 [0011] The details of the present invention are described below.
- [0012] In the lubricating oil composition of the present invention, the component herein identified by (A) is a mineral oil-derived lubricating base oil containing 80% by mass or more of a saturated hydrocarbon component which has a viscosity-density constant of 0.79 or less.
- 40 [0013] Component (A) contains a saturated hydrocarbon component in an amount of more than 80% by mass, preferably more than 90% by mass (90 to 100% by mass), and more preferably more than 95% by mass (95 to 100% by mass).
- [0014] Component (A) containing less than 80% by mass of a saturated hydrocarbon component is not preferred because it fail to provide a lubricating oil composition having a sufficient oxidation stability.
- 45 [0015] The term "content of a saturated hydrocarbon component" as referred to herein designates a mass percentage of the saturated hydrocarbon component separated with respect to the total amount of the sample, wherein the mass percentage is measured in accordance with silica/alumina gel chromatography described in "Separation of High-Boiling Petroleum Distillate Using Gradient Elution Through Dual-Packed (Silica Gel-Alumina Gel) Adsorption Columns" described in Analytical Chemistry, Vol., 44, No. 6 (1972), p. 915-919, except that n-pentane as used therein for use in
- 50 the elution of the saturated hydrocarbon component is replaced with n-hexane.
- [0016] Further, the saturated hydrocarbon component, which is separated in accordance with the above-mentioned method and which is contained in Component (A), needs to have a viscosity-density constant of 0.79 or less, and preferably 0.785 or less.
- [0017] The viscosity-density constant of the saturated hydrocarbon component exceeding 0.79 is not preferred because the resulting lubricating oil composition has a poor oxidation stability. Meanwhile, although the lower limit of the viscosity-density constant of the saturated hydrocarbon component is not limited, it is usually 0.76 or more.
- 55 [0018] The viscosity-density constant as referred to herein designates the value which is obtained in accordance with the method for calculating a viscosity-density constant described in ASTM D2140 "Standard Test Method for Carbon-

Type Composition of Insulating Oils of Petroleum Origin", except that the specific gravity at 60° F(15.6°C) is replaced with the density at 15°C and the kinetic viscosity at 100° F(37.8°C), is replaced with the kinetic viscosity at 40°C.

[0019] That is, the viscosity-density constant as referred to herein means the value which is derived from the following equation :

$$\text{viscosity-density constant} = \{G + 0.0887 - 0.776 \log(\log(10V-4))\} / [1.082 - 0.72 \log(\log(10V-4))]$$

(a)

where G is the density (g/cm³) at 15°C and V is the kinetic viscosity (mm²/s) at 40°C.

[0020] The method for producing the mineral oil-derived lubricating base oil identified by Component (A) of the present invention is not particularly limited. For example, the lubricating base oil eligible for the purpose of the present invention may be a paraffinic oil or a naphthenic oil obtained by subjecting a lubricating oil fraction derived from an atmospheric or vacuum distillation of crude oil to one or more of refining processes such as solvent deasphalting, solvent extraction, hydrocracking, solvent dewaxing, catalytic dewaxing, hydrotreating, washing with sulfuric acid treatment with clay, and the like. These base oils may be used independently or in a combination of two or more of them at any blending ratio.

[0021] Further, Component (A) as defined herein is preferably obtained in the following procedures. That is, for example, one of the following oils is used as a feedstock oil.

- (1) a distillate derived from the atmospheric distillation of a paraffinic crude oil and/or a mixed crude oil;
- (2) a whole vacuum gas oil (WVGO) of a paraffinic crude oil and/or a mixed crude oil;
- (3) an oil obtained by subjecting (1) and/or (2) to mild hydrocracking (MHC);
- (4) a mixture of two or more oils selected from (1) to (3);
- (5) a deasphalted oil (DAO) of (1), (2), (3) or (4);
- (6) an oil obtained by subjecting (5) to mild hydrocracking (MHC); and
- (7) a mixture of two or more oils selected from (1) to (6).

[0022] Either the feedstock oil itself or a lubricating oil fraction recovered therefrom is refined by a conventional refining process to obtain a lubricating oil fraction which serves as Component (A) of the invention.

[0023] The conventional refining process may be any refining process which is usually employed for the production of a base oil. Examples of the conventional refining process as referred to herein include (1) hydrotreating such as hydrocracking or hydrofinishing, (2) solvent refining such as solvent extraction with use of furfural, (3) dewaxing such as solvent dewaxing or catalytic dewaxing, (4) clay treatment with use of acid clay or activated clay, and (5) chemical refining with use of a chemical (acid or alkali) such as washing with sulfuric acid or washing with caustic soda. Any one of these processes or a combination of two or more of these processes in any order can be adopted in the present invention.

[0024] In particular, the mineral oil-derived lubricating base oil defined as Component (A) in the present invention desirably contains, based on the total amount thereof, a substance in an amount of preferably more than 50% by mass, more preferably more than 70% by mass, and most preferably more than 80% by mass, which substance is produced by hydrocracking a feedstock oil selected from the above-described oils (1) through (7) as it is or the lubricating oil fractions recovered therefrom and then subjecting the resulting product or the lubricating oil fraction recovered therefrom to dewaxing such as solvent dewaxing and catalytic dewaxing and then solvent refining, alternatively followed by the further dewaxing.

[0025] Although the kinetic viscosity at 40°C of the component (A) for use in the present invention is not particularly limited, the kinetic viscosity at 40°C is preferably 5 mm²/s or more, and more preferably 10 mm²/s or more. Further, the kinetic viscosity at 40°C is preferably 200 mm²/s or less and more preferably 100 mm²/s or less.

[0026] The use of Component (A) having the kinetic viscosity at 40°C of 5 mm²/s or more results in the production of a lubricating oil composition which is satisfactory in oil film forming performance and excels in lubricity as well as reduced in the evaporation loss of the base oil at a high temperature. Meanwhile, the kinetic viscosity at 40°C of 200 mm²/s or less leads to a reduction in fluid resistance, resulting in a lubricating oil having a less frictional resistance at lubricated site.

[0027] Although Component (A) is not particularly limited on the kinetic viscosity at 100°C, it is preferably 0.5 mm²/s or more, and more preferably 1 mm²/s or more. Furthermore, the kinetic viscosity at 100°C is preferably 100 mm²/s or

less and more preferably 50 mm²/s or less.

[0028] The use of Component (A) having the kinetic viscosity at 100°C of 5 mm²/s or more results in a lubricating oil composition which is satisfactory in oil film forming performance and excels in lubricity as well as reduced in the evaporation loss of the base oil at a high temperature. Meanwhile, the kinetic viscosity at 100°C of 100 mm²/s or less leads to a reduction in fluid resistance, resulting in the production of a lubricating oil having a less frictional resistance at lubricated site.

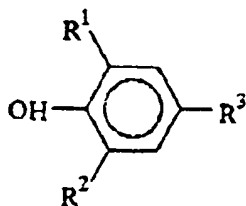
[0029] Although there is no particular limitation imposed on the viscosity index of Component (A), it is preferably 50 or more and more preferably 80 or more. The viscosity index of 50 or more leads to a lubricating oil composition having better-consistent oil film forming and fluid resistance reducing performances.

[0030] Although the pour point of Component (A) is not particularly limited, it is preferably 0°C or lower and more preferably -5°C or lower. The pour point of 0°C or lower of Component (A) leads to a lubricating oil composition which is less likely to hinder the operation of a machine at a low temperature.

[0031] The oxidation inhibitor identified herein by Component (B) contained in the lubricating oil composition of the present invention is not particularly limited. For example, the oxidation inhibitor eligible for the purpose of the invention is at least one oxidation inhibitor selected from the group consisting of a hindered phenol compound, an aromatic amine compound and a sulfur containing compound.

[0032] The hindered phenol-based oxidation inhibitor may be any hindered phenol compound as long as it can be utilized as an oxidation inhibitor for a lubricating oil and thus is not limited to a particular type.

[0033] More specifically, preferred examples of such a hindered phenol compound include a compound or a mixture of two or more compounds selected from those represented by the following formula

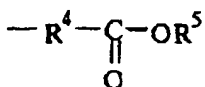


(I)

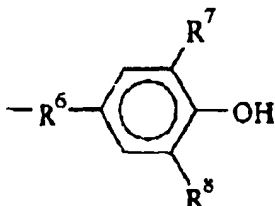
wherein R¹ and R² are independently an alkyl group having 1 to 4 carbon atoms with the proviso that at least either R¹ or R² is a tert-butyl group.

[0034] R¹ and R² may be methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl and tert-butyl. However, one of R¹ and R² is preferably tert-butyl while the other is preferably methyl, ethyl or tert-butyl.

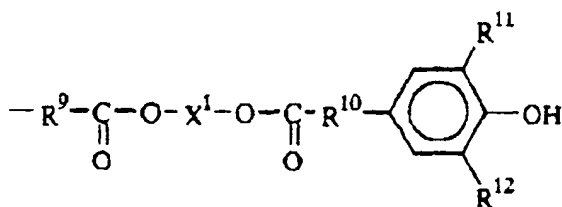
[0035] In formula (I), R³ is an alkyl group having 1 to 4 carbon atoms or a group selected from groups represented by the following formulae



(II)



(III) and



(IV)

[0036] Examples of the alkyl group represented by R^3 include methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl and tert-butyl, among which methyl and ethyl are preferred in view of excellent oxidation stability.

[0037] Particularly preferred examples of the hindered phenol compound represented by formula (I) where R^3 is an alkyl group having 1 to 4 carbon atoms include 2,6-di-tert-butyl-p-cresol, 2,6-di-tert-butyl-4-ethylphenol and a mixture thereof.

[0038] In the group represented by formula (II), R^4 is an alkylene group having 1 to 6 carbon atoms, while R^5 is an alkyl group having 1 to 24 carbon atoms or an alkenyl group.

[0039] The alkylene group represented by R^4 may be straight or branched. Specific examples include methylene, methylenemethylene, ethylene (dimethylene), ethylmethylene, propylene (methylethylene), trimethylene, straight or branched butylene, straight or branched pentylene and straight or branched hexylene. Among these groups, alkylene groups having 1 to 2 carbon atoms such as methylene, methylenemethylene and ethylene (dimethylene) are more preferred.

[0040] The alkyl group or the alkenyl group each represented by R^5 may be straight or branched.

[0041] Specific examples of R^5 include alkyl groups such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, straight or branched pentyl, straight or branched hexyl, straight or branched heptyl, straight or branched octyl, straight or branched nonyl, straight or branched decyl, straight or branched undecyl, straight or branched dodecyl, straight or branched tridecyl, straight or branched tetradecyl, straight or branched pentadecyl, straight or branched hexadecyl, straight or branched heptadecyl, straight or branched octadecyl, straight or branched nonadecyl, straight or branched icosyl, straight or branched heneicosyl, straight or branched docosyl, straight or branched tricosyl and straight or branched tetracosyl and alkenyl groups such as vinyl, propenyl, isopropenyl, straight or branched butenyl, straight or branched pentenyl, straight or branched hexenyl, straight or branched heptenyl, straight or branched octenyl, straight or branched nonenyl, straight or branched decenyl, straight or branched undecenyl, straight or branched dodecenyl, straight or branched tridecenyl, straight or branched tetradecenyl, straight or branched pentadecenyl, straight or branched hexadecenyl, straight or branched heptadecenyl, straight or branched octadecenyl, straight or branched octadecadienyl, straight or branched nonadecenyl, straight or branched icosenyl, straight or branched heneicosenyl, straight

or branched docosenyl, straight or branched tricosenyl and straight or branched tetracosenyl.

[0042] From the standpoint of better solubility in the lubricating base oil, preferred alkyl groups represented by R^5 are those having 4 to 18 carbon atoms such as n-butyl, isobutyl, sec-butyl, tert-butyl, straight or branched pentyl, straight or branched hexyl, straight or branched heptyl, straight or branched octyl, straight or branched nonyl, straight or branched decyl, straight or branched undecyl, straight or branched dodecyl, straight or branched tridecyl, straight or branched tetradecyl, straight or branched pentadecyl, straight or branched hexadecyl, straight or branched heptadecyl and straight or branched octadecyl. Among these groups, straight or branched alkyl groups having 6 to 12 carbon atoms are more preferred and branched alkyl groups having 6 to 12 carbon atoms are particularly preferred.

[0043] Among the groups represented by formula (II), more preferred are those of the formula wherein R^4 is an alkylene group having 1 to 2 carbon atoms, while R^5 is a straight or branched alkyl group having 6 to 12 carbon atoms; and most preferred are those of the formula wherein R^4 is an alkylene group having 1 to 2 carbon atoms, while R^5 is a branched alkyl group having 6 to 12 carbon atoms.

[0044] Particularly preferred hindered phenols represented by formula (I) where R^3 is a group represented by formula (II) are

15 n-hexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 isoheptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 n-heptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 isoheptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 20 n-octyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 isooctyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 2-ethylhexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 n-nonyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 isononyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 25 n-decyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 isodecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 n-undecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 isoundecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 n-dodecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 30 isododecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate,
 n-hexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 isoheptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 n-heptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 isoheptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 35 n-octyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 isooctyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 2-ethylhexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 n-nonyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 isononyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 40 n-decyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 isodecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 n-undecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 isoundecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 n-dodecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 45 isododecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate,
 n-hexyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 isoheptyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 n-heptyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 isoheptyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 50 n-octyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 isooctyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 2-ethylhexyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 n-nonyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 isononyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 55 n-decyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 isodecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 n-undecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 isoundecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,

n-dodecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 isododecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate,
 n-hexyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 isohexyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 5 n-heptyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 isoheptyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 n-octyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 isooctyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 2-ethylhexyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 10 n-nonyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 isononyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 n-decyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 isodecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 n-undecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 15 isoundecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 n-dodecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,
 isododecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate and a mixture thereof.

[0045] In the group represented by formula (III), R^6 is an alkylene group having 1 to 6 carbon atoms, while R^7 and R^8 are independently an alkyl group having 1 to 4 carbon atoms with the proviso that at least either R^7 or R^8 is tert-butyl.

[0046] The alkylene group represented by R^6 may be straight or branched. Specific examples of R^6 include the alkylene groups previously enumerated for R^4 . Among these groups, alkylene groups having 1 to 2 carbon atoms such as methylene, methylenemethylene and ethylene (dimethylene) are more preferred.

[0047] Meanwhile, the alkyl groups represented by R^7 and R^8 each may be straight or branched. Specific examples include the alkyl groups previously enumerated for R^1 and R^2 , respectively. Among these groups, preferred is the combination where one of R^7 and R^8 is tert-butyl, while the other is methyl, ethyl or tert-butyl.

[0048] As for the groups represented by formula (III), particularly preferred are those where R^6 is an alkylene group having 1 or 2 carbon atoms, while either R^7 or R^8 is tert-butyl, and the other is methyl, ethyl or tert-butyl.

[0049] Particularly preferred hindered phenols represented by formula (I) where R^3 is a group represented by formula (III) are

bis(3,5-di-tert-butyl-4-hydroxyphenyl)methane,
 1,1-bis(3,5-di-tert-butyl-4-hydroxyphenyl)ethane,
 1,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)ethane,
 35 1,1-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane,
 1,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane,
 1,3-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane,
 2,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane and a mixture thereof.

[0050] In the groups represented by formula (IV), R^9 and R^{10} are independently an alkylene group having 1 to 6 carbon atoms; R^{11} and R^{12} are independently an alkyl group having 1 to 4 carbon atoms with the proviso that at least one of R^{11} and R^{12} is a tert-butyl group, and X^1 is either an alkylene group having 1 to 18 carbon atoms or a group represented by the formula



(V)

50 where R^{13} and R^{14} are independently an alkylene group having 1 to 6 carbon atoms.

[0051] The alkyl groups represented by R^9 and R^{10} may be straight or branched. Specific examples include the alkylene groups previously enumerated for R^4 . More specifically, R^9 and R^{10} are independently an alkylene group having 1 or 2 carbon atoms, preferably methylene, methylenemethylene, and ethylene (dimethylene).

55 [0052] Meanwhile, the alkyl groups represented by R^{11} and R^{12} may be straight or branched. Specific examples include the alkyl groups previously enumerated for R^1 and R^2 , respectively. Among these groups, one of R^{11} and R^{12} is preferably tert-butyl, and the other is preferably methyl, ethyl or tert-butyl.

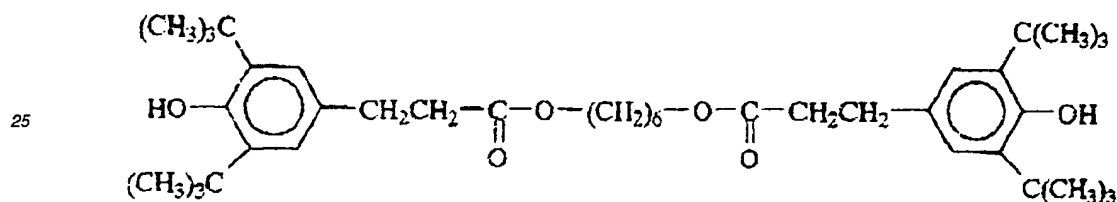
[0053] Specific examples of the alkylene group represented by X^1 include methylene, methylenemethylene, ethylene

(dimethylene), ethylmethylene, propylene (methylethylene), trimethylene, straight or branched butylene, straight or branched pentylene, straight or branched hexylene, straight or branched heptylene, straight or branched octylene, straight or branched nonylene, straight or branched decylene, straight or branched undecylene, straight or branched dodecylene, straight or branched tridecylene, straight or branched tetradecylene, straight or branched pentadecylene, straight or branched hexadecylene, straight or branched heptadecylene and straight or branched octadecylene. Among these groups, preferred are alkylene groups having 1 to 6 carbon atoms such as methylene, methylmethylene, ethylene (dimethylene), ethylmethylene, propylene (methylethylene), trimethylene straight or branched butylene, straight or branched pentylene and straight or branched hexylene, and particularly preferred are alkylene groups having a straight chain of 2 to 6 carbon atoms such as ethylene (dimethylene), trimethylene, straight butylene (tetramethylene) straight pentylene (pentamethylene) and straight hexylene (hexamethylene).

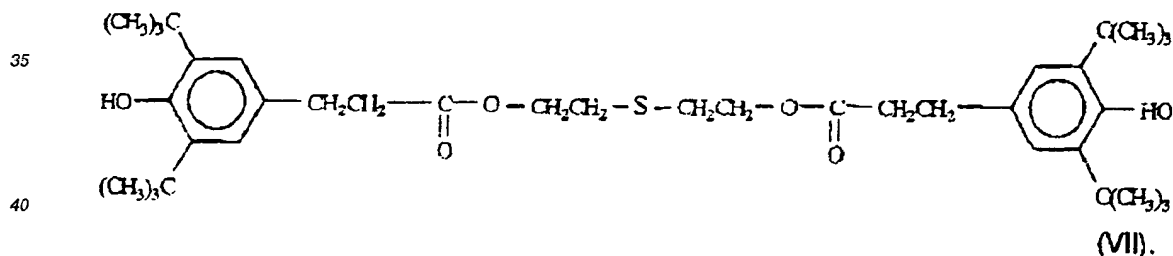
[0054] In formula (V) for X^1 , the alkylene group represented by R^{13} and R^{14} may be straight or branched. Specific examples include the alkylene groups previously enumerated for R^4 . Among these groups, alkylene groups having 1 or 2 carbon atoms such as methylene, methylmethylene and ethylene (dimethylene) are more preferred.

[0055] In the group represented by formula (IV), most preferably R^9 and R^{10} are independently an alkylene group having 1 or 2 carbon atoms; one of R^{11} and R^{12} is tert-butyl and the other is methyl, ethyl or tert-butyl; and X^1 is either a straight alkylene group having 2 to 6 carbon atoms or a group represented by formula (IV) where R^{13} and R^{14} are independently an alkylene group having 1 or 2 carbon atoms.

[0056] Examples of the most preferred hindered phenol compound represented by formula (I) where R^3 is a group represented by formula (IV) include compounds represented by the following formulae:

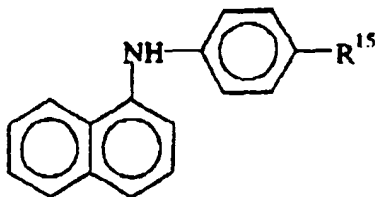


(VI) and



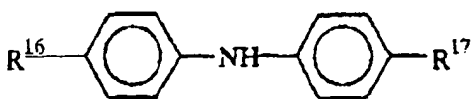
[0057] The aromatic amine-based oxidation inhibitor which is also one of Components (B) for the lubricating oil composition of the present invention may be any aromatic amine as long as it is eligible for an oxidation inhibitor for lubrication oils and thus is not particularly limited.

[0058] Preferred examples of such a compound include one compound or a mixture of two or more compounds selected from phenyl- α -naphthylamine, N-alkylphenyl- α -naphthylamine represented by the formula



(VIII),

and p, p'-dialkyldiphenylamine represented by the formula



(IX).

[0059] In formula (VIII), R^{15} is a straight or branched alkyl group having 1 to 16 carbon atoms.

[0060] Specific examples of R^{15} include alkyl groups such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, straight or branched pentyl, straight or branched hexyl, straight or branched heptyl, straight or branched octyl, straight or branched nonyl, straight or branched decyl, straight or branched undecyl, straight or branched dodecyl, straight or branched tridecyl, straight or branched tetradecyl, straight or branched pentadecyl and straight or branched hexadecyl.

[0061] Among these groups, because of better solubility of the oxidation product of the oxidation inhibitor itself in the lubricating base oil, R^{15} is preferably a branched alkyl group having 8 to 16 carbon atoms and more preferably a branched alkyl group having 8 to 16 carbon atoms derived from an oligomer of olefins having 3 or 4 carbon atoms.

[0062] Specific examples of the olefins having 3 or 4 carbon atoms are propylene, 1-butene, 2-butene and isobutylene. Among these olefins, because of better solubility of the oxidation product of the oxidation inhibitor itself in the lubricating base oil, propylene and isobutylene are preferred, and propylene is more preferred.

[0063] More specifically, particularly preferred for R^{15} are a branched octyl group derived from a dimer of isobutylene, a branched nonyl group derived from a trimer of propylene, a branched dodecyl group derived from a trimer of isobutylene, a branched dodecyl group derived from a tetramer of propylene and a branched pentadecyl group derived from a pentamer of propylene. Most preferred are a branched dodecyl group derived from a tetramer of propylene and a branched pentadecyl group derived from a pentamer of propylene.

[0064] In formula (IX), R^{16} and R^{17} are independently a straight or branched alkyl group having 1 to 16 carbon atoms.

[0065] Examples of R^{16} and R^{17} include the alkyl groups previously enumerated for R^{15} . Among these groups, because of better solubility of the oxidation product of the oxidation inhibitor itself in the lubricating base oil, R^{16} and R^{17} each are preferably a branched alkyl group having 8 to 16 carbon atoms. More preferably R^{16} and R^{17} are independently a branched alkyl group having 8 to 16 carbon atoms and derived from an oligomer of olefins having 3 or 4 carbon atoms.

[0066] Specific examples of the olefins having 3 or 4 carbon atoms are example propylene, 1-butene, 2-butene and isobutylene. Among these olefins, because of better solubility of the oxidation product of the oxidation inhibitor itself in the lubricating base oil, propylene and isobutylene are preferred and propylene is more preferred.

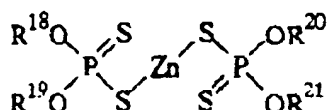
[0067] More specifically, particularly preferred for each R^{16} and R^{17} are a branched octyl group derived from a dimer of isobutylene, a branched nonyl group derived from a trimer of propylene, a branched dodecyl group derived from a trimer of isobutylene, a branched dodecyl group derived from a tetramer of propylene, or a branched pentadecyl group derived from a pentamer of propylene, among which a branched nonyl group derived from a trimer of propylene, a branched dodecyl group derived from a tetramer of propylene and a branched pentadecyl group derived from a pen-

tamer of propylene are most preferred.

[0068] The sulfur-based oxidation inhibitor which is also one of Components (B) in the lubricating oil composition of the present invention may be any sulfur containing compound which can be used as an oxidation inhibitor for lubricating oils and is thus not particularly limited.

- 5 [0069] Preferred examples of such a compound include one compound or a mixture of two or more compounds selected from metal salts of dihydrocarbyldithiophosphoric acid such as zinc dihydrocarbyldithiophosphate and molybdenum dihydrocarbyldithiophosphate, metal salts of dithiocarbamic acid such as zinc dihydrocarbyldithiocarbamate and copper dihydrocarbyldithiocarbamate and sulfides. Among these compounds, zinc dihydrocarbyldithiophosphate, zinc dihydrocarbyldithiocarbamate molybdenum dihydrocarbyldithiocarbamate and sulfides are more preferred.

- 10 [0070] Preferred examples of the zinc dihydrocarbyldithiophosphate suitable for the sulfur-containing oxidation inhibitor are compounds represented by the formula



(X).

- [0071] In formula (X), R¹⁸, R¹⁹, R²⁰ and R²¹ are independently a hydrocarbon group having 1 to 18 carbon atoms. Examples of such a hydrocarbon group include alkyl groups having 1 to 18 carbon atoms such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, straight or branched pentyl straight or branched hexyl, straight or branched heptyl, straight or branched octyl, straight or branched nonyl, straight or branched decyl, straight or branched undecyl, straight or branched dodecyl, straight or branched tridecyl, straight or branched tetradecyl, straight or branched pentadecyl, straight or branched hexadecyl straight or branched heptadecyl and straight or branched octadecyl; alkenyl groups having 4 to 18 carbon atoms such as straight or branched butenyl, straight or branched pentenyl, straight or branched hexenyl, straight or branched heptenyl, straight or branched octenyl, straight or branched nonenyl, straight or branched decenyl, straight or branched undecenyl, straight or branched dodecenyl, straight or branched tri-
 30 decenyl, straight or branched tetradecenyl, straight or branched pentadecenyl, straight or branched hexadecenyl, straight or branched heptadecenyl and straight or branched octadecenyl; cycloalkyl groups having 5 to 7 carbon atoms such as cyclopentyl, cyclohexyl and cycloheptyl; alkylcycloalkyl groups having 6 to 11 carbon atoms such as methylcyclopentyl, dimethylcyclopentyl (inclusive of all structural isomers thereof), methylethylcyclopentyl (inclusive of all structural isomers thereof), diethylcyclopentyl (inclusive of all structural isomers thereof), methylcyclohexyl, dimethylcyclohexyl (inclusive of all structural isomers thereof), methylethylcyclohexyl (inclusive of all structural isomers thereof), diethylcyclohexyl (inclusive of all structural isomers thereof), methylcycloheptyl, dimethylcycloheptyl (inclusive of all structural isomers thereof), methylethylcycloheptyl (inclusive of all structural isomers thereof) and diethylcycloheptyl (inclusive of all structural isomers thereof); aryl groups such as phenyl and naphthyl; alkylaryl groups having 7 to 18 carbon atoms such as tolyl (inclusive of all structural isomers thereof), xylyl (inclusive of all structural isomers thereof), ethylphenyl (inclusive of all structural isomers thereof), straight or branched propylphenyl (inclusive of all structural isomers thereof), straight or branched butylphenyl (inclusive of all structural isomers thereof), straight or branched pentylphenyl (inclusive of all structural isomers thereof), straight or branched hexylphenyl (inclusive of all structural isomers thereof), straight or branched heptylphenyl (inclusive of all structural isomers thereof), straight or branched octylphenyl (inclusive of all structural isomers thereof), straight or branched nonylphenyl (inclusive of all structural isomers thereof), straight or branched decylphenyl (inclusive of all structural isomers thereof), straight or branched undecylphenyl (inclusive of all structural isomers thereof) and straight or branched dodecylphenyl (inclusive of all structural isomers thereof); and arylalkyl groups having 7 to 12 carbon atoms such as benzyl, phenylethyl, phenylpropyl (inclusive of isomers of propyl group), phenylbutyl (inclusive of isomers of butyl group), phenylpentyl (inclusive of isomers of pentyl group) and phenylhexyl (inclusive of isomers of hexyl group).

- [0072] The above-mentioned alkyl groups and alkenyl groups may be so-called primary alkyl groups and alkenyl groups in which the carbon atom bonded to an oxygen atom is a primary carbon atom, may be so-called secondary alkyl groups and alkenyl groups in which the carbon atom bonded to an oxygen atom is a secondary carbon atom, or may be so-called tertiary alkyl groups and alkenyl groups in which the carbon atom bonded to an oxygen atom is a tertiary carbon atom.

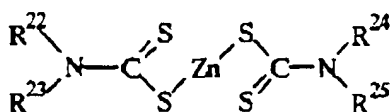
[0073] Among the compounds represented by formula (X), the zinc dialkyldithiophosphates in which R¹⁸, R¹⁹, R²⁰ and R²¹ each are a straight or branched hydrocarbon group having 1 to 18 carbon atoms are preferred because this

compound provides a lubricating oil composition particularly excelled in an oxidation inhibition properties.

[0074] Specific examples of the zinc dihydrocarbyldithiophosphates include

5 zinc diethyldithiophosphate, zinc di-n-propyldithiophosphate,
zinc diisopropyldithiophosphate, zinc di-n-butyldithiophosphate,
zinc diisobutyldithiophosphate, zinc di-sec-butyldithiophosphate,
zinc di-tert-butyldithiophosphate,
zinc di(straight or branched pentyl)dithiophosphate,
zinc di(straight or branched hexyl)dithiophosphate,
10 zinc di(straight or branched heptyl)dithiophosphate,
zinc di(straight or branched octyl)dithiophosphate,
zinc di(straight or branched nonyl)dithiophosphate,
zinc di(straight or branched decyl)dithiophosphate,
zinc di(straight or branched undecyl)dithiophosphate,
15 zinc di(straight or branched dodecyl)dithiophosphate,
zinc di(straight or branched tridecyl)dithiophosphate,
zinc di(straight or branched tetradecyl)dithiophosphate,
zinc di(straight or branched pentadecyl)dithiophosphate,
zinc di(straight or branched hexadecyl)dithiophosphate,
20 zinc di(straight or branched octadecyl)dithiophosphate,
zinc diphenyldithiophosphate, zinc diethylphenyldithiophosphate,
zinc dipropylphenyldithiophosphate, zinc diisopropylphenyldithiophosphate,
zinc dibutylphenyldithiophosphate, zinc dipentylphenyldithiophosphate,
zinc dihexylphenyldithiophosphate, zinc diheptylphenyldithiophosphate,
25 zinc dioctylphenyldithiophosphate, zinc di-2-ethylhexylphenyldithiophosphate,
zinc dinonylphenyldithiophosphate, zinc didecylphenyldithiophosphate,
zinc diundecylphenyldithiophosphate, zinc didodecylphenyldithiophosphate,
zinc ditridecylphenyldithiophosphate, zinc ditetradecylphenyldithiophosphate,
zinc dipentadecylphenyldithiophosphate,
30 zinc dihexadecylphenyldithiophosphate,
zinc diheptadecylphenyldithiophosphate, zinc dioctadecylphenyldithiophosphate, and a mixture thereof.

[0075] Preferred examples of the zinc dihydrocarbyldithiocarbamates exemplified as another suitable sulfur-containing oxidation inhibitor are compounds represented by the formula



(XI)

45 [0076] In formula (XI), R^{22} , R^{23} , R^{24} and R^{25} are independently a hydrocarbon group having 1 to 18 carbon atoms.

[0077] Specific examples of R^{22} , R^{23} , R^{24} and R^{25} are alkyl groups, alkenyl groups, cycloalkyl groups, alkylcycloalkyl groups, aryl groups, alkylaryl groups, and arylalkyl groups which are previously exemplified with respect to the hydrocarbon groups represented by R^{18} , R^{19} , R^{20} or R^{21} .

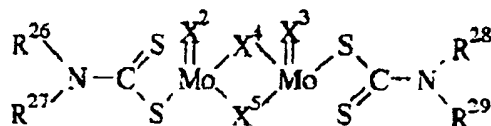
50 [0078] The above-mentioned alkyl groups and alkenyl groups may be so-called primary alkyl groups and alkenyl groups in which the carbon atom bonded to an oxygen atom is a primary carbon atom, may be so-called secondary alkyl groups and alkenyl groups in which the carbon atom bonded to an oxygen atom is a secondary carbon atom, or may be so-called tertiary alkyl groups and alkenyl groups in which the carbon atom bonded to an oxygen atom is a tertiary carbon atom.

55 [0079] Among the compounds represented by formula (XI), the zinc dialkyldithiocarbamates in which R^{22} , R^{23} , R^{24} and R^{25} are independently an alkyl group having 4 to 13 carbon atoms are preferred because the compounds provide a lubricating oil composition with a particularly high oxidation stability.

[0080] Preferred specific examples of the zinc dihydrocarbyldithiocarbamates include zinc di(straight or branched)butyldithiocarbamate,

zinc di(straight or branched)pentylthiocarbamate,
 zinc di(straight or branched)hexylthiocarbamate,
 zinc di(straight or branched)heptylthiocarbamate,
 zinc di(straight or branched)octylthiocarbamate,
 zinc di(straight or branched)nonylthiocarbamate,
 zinc di(straight or branched)decylthiocarbamate,
 zinc di(straight or branched)undecylthiocarbamate,
 zinc di(straight or branched)dodecylthiocarbamate,
 zinc di(straight or branched)tridecylthiocarbamate,
 zinc di(straight or branched)tetradecylthiocarbamate and a mixture thereof.

[0081] Preferred examples of the molybdenum dihydrocarbyldithiocarbamates which are another examples of the sulfur-containing oxidation inhibitor are compounds represented by the formula



(XII).

[0082] In formula (XII), R^{26} , R^{27} , R^{28} and R^{29} are independently a hydrocarbon group having 1 to 18 carbon atoms.

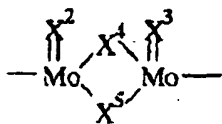
[0083] Specific examples of R^{26} , R^{27} , R^{28} and R^{29} are alkyl groups, alkenyl groups, cycloalkyl groups, alkylcycloalkyl groups, aryl groups, alkylaryl groups, and arylalkyl groups which are previously exemplified with respect to the hydrocarbon groups represented by R^{18} , R^{19} , R^{20} or R^{21} .

[0084] The above-mentioned alkyl groups and alkenyl groups may be so-called primary alkyl groups and alkenyl groups in which the carbon atom bonded to an oxygen atom is a primary carbon atom, may be so-called secondary alkyl groups and alkenyl groups in which the carbon atom bonded to an oxygen atom is a secondary carbon atom, or may be so-called tertiary alkyl groups and alkenyl groups in which the carbon atom bonded to an oxygen atom is a tertiary carbon atom.

[0085] Among the compounds represented by formula (XII), the molybdenum dialkylthiocarbamates in which R^{26} , R^{27} , R^{28} and R^{29} are independently an alkyl group having 4 to 13 carbon atoms are preferred, because the compounds provide a lubricating oil composition with a particularly high oxidation stability.

[0086] The designations X^2 , X^3 , X^4 and X^5 inclusive in formula (XII) each are a sulfur atom or an oxygen atom, but at least one such atom should be a sulfur atom with a view to achieving an improved oxidation stability.

[0087] In the present invention, two or more of the molybdenum dialkylthiocarbamates of (XII) may be used in combination at any ratio therebetween, in which instance the atom group of the formula



(XIII)

may be represented in its average structure represented by $-Mo_2S_aO_{(4-a)}-$ where a is a numeral preferably in the range of 1 to 3 and more preferably in the range of 1.5 to 2.5 with a view to ensuring oxidation stability and corrosion-resist-

ance at bearings and other mechanical parts of the engines.

[0088] X^2 , X^3 , X^4 and X^5 in the group of atoms in formula (XIII) correspond to X^2 , X^3 , X^4 and X^5 in formula (XII), respectively.

[0089] Particularly preferred examples of the molybdenum dihydrocarbyldithiocarbamates include

5

molybdenum sulfide di(straight or branched)butyldithiocarbamate,
 molybdenum sulfide di(straight or branched)pentyldithiocarbamate,
 molybdenum sulfide di(straight or branched)hexyldithiocarbamate,
 molybdenum sulfide di(straight or branched)heptyldithiocarbamate,
 10 molybdenum sulfide di(straight or branched)octyldithiocarbamate,
 molybdenum sulfide di(straight or branched)nonyldithiocarbamate,
 molybdenum sulfide di(straight or branched)decyldithiocarbamate,
 molybdenum sulfide di(straight or branched)undecyldithiocarbamate,
 molybdenum sulfide di(straight or branched)dodecyldithiocarbamate,
 15 molybdenum sulfide di(straight or branched)tridecyldithiocarbamate,
 molybdenum sulfide di(straight or branched)butyldithiocarbamate,
 molybdenum sulfide di(straight or branched)pentyldithiocarbamate,
 molybdenum sulfide di(straight or branched)hexyldithiocarbamate,
 molybdenum sulfide di(straight or branched)heptyldithiocarbamate,
 20 molybdenum sulfide di(straight or branched)octyldithiocarbamate,
 molybdenum sulfide di(straight or branched)nonyldithiocarbamate,
 molybdenum sulfide di(straight or branched)decyldithiocarbamate,
 molybdenum sulfide di(straight or branched)undecyldithiocarbamate,
 molybdenum sulfide di(straight or branched)dodecyldithiocarbamate,
 25 molybdenum sulfide di(straight or branched)tridecyldithiocarbamate and a mixture thereof.

[0090] Preferred examples of the sulfides exemplified as another suitable sulfur-containing oxidation inhibitors are dihydrocarbyl sulfides represented by the formula

30



35 **[0091]** In formula (XIV), R³⁰ and R³¹ are independently selected from a straight or branched alkyl group having 1 to 22 carbon atoms; an aryl group having 6 to 20 carbon atoms, an alkylaryl group having 7 to 20 carbon atoms and an arylalkyl group having 7 to 20 carbon atoms; b is a numeral of 1 to 5, more preferably 1 or 2, and most preferably 2.

[0092] Examples of R³⁰ and R³¹ include independently alkyl groups such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, straight or branched pentyl, straight or branched hexyl, cyclohexyl, straight or branched heptyl, straight or branched octyl (e.g., tert-octyl), straight or branched nonyl, straight or branched decyl, straight or branched undecyl, straight or branched dodecyl (e.g., tert-dodecyl), straight or branched tridecyl, straight or branched tetradecyl, straight or branched pentadecyl, straight or branched hexadecyl (e.g., 2-methylpentadecyl and tert-hexadecyl), straight or branched heptadecyl, straight or branched octadecyl, straight or branched nonadecyl, straight or branched icosyl, straight or branched henicosyl and straight or branched docosyl; alkenyl groups such as propenyl and butenyl; aryl groups such as phenyl and naphthyl(including all isomers); alkylaryl groups (in which the alkyl portion may be straight or branched and may be positioned in any substitution site on the aryl group) such as tolyl, ethylphenyl, propylphenyl, butylphenyl, pentylphenyl, hexylphenyl, heptylphenyl, octylphenyl, nonylphenyl, decylphenyl, undecylphenyl, dodecylphenyl, xylyl, ethylmethylphenyl, diethylphenyl, dipropylphenyl, dibutylphenyl, methyl-naphthyl, ethyl-naphthyl, propyl-naphthyl, butyl-naphthyl dimethyl-naphthyl, ethylmethyl-naphthyl, diethyl-naphthyl, dipropyl-naphthyl and dibutyl-naphthyl; and arylalkyl groups (in which the alkyl portion may be straight or branched and the aryl portion may be positioned in any substitution site) such as benzyl, phenethyl and phenylpropyl.

[0093] Among these groups represented by R³⁰ and R³¹, preferred are alkyl groups having 3 to 18 carbon atoms and derived from propylene or isobutene, aryl groups having 6 to 8 carbon atoms, alkylaryl groups having 7 to 8 carbon atoms and arylalkyl groups having 7 to 8 carbon atoms.

55 **[0094]** More specifically, examples of these groups include alkyl groups (inclusive of all branched isomers thereof) such as an isopropyl group, a branched hexyl group derived from a dimer of propylene, a branched nonyl group derived from a trimer of propylene, a branched dodecyl group derived from a tetramer of propylene, a branched pentadecyl group derived from a pentamer of propylene, a branched octadecyl group derived from a hexamer of propylene, a tert-

butyl group, a branched octyl group derived from a dimer of isobutene, a branched dodecyl group derived from a trimer of isobutene and a branched hexadecyl group derived from a tetramer of isobutene; alkenyl groups such as propenyl and butenyl; alkylaryl groups (in which the alkyl portion may be straight or branched and may be positioned in any substitution site on the aryl group) such as phenyl, tolyl, ethylphenyl and xylyl; and arylalkyl groups (in which the phenyl group may be positioned in any substitution site) such as benzyl and phenylethyl. Among these groups, preferred are alkyl groups having 3 to 18 carbon atoms, preferably 6 to 15 carbon atoms and derived from propylene or isobutene.

[0095] Specific examples of the dihydrocarbylsulfides include dimethyl sulfides such as dimethyl monosulfide and dimethyl disulfide; diethyl sulfides such as diethyl monosulfide and diethyl disulfide; di-n-propyl sulfides such as di-n-propyl monosulfide and di-n-propyl disulfide; diisopropyl sulfides such as diisopropyl monosulfide and diisopropyl disulfide; di-n-butyl sulfides such as di-n-butyl monosulfide and di-n-butyl disulfide; diisobutyl sulfides such as diisobutyl monosulfide and diisobutyl disulfide; di-sec-butyl sulfides such as di-sec-butyl monosulfide and di-sec-butyl disulfide; di-tert-butyl sulfides such as di-tert-butyl monosulfide and di-tert-butyl disulfide; di(straight or branched pentyl) sulfides such as di(straight or branched pentyl) monosulfide and di(straight or branched pentyl) disulfide; di(straight or branched hexyl) sulfides such as di(straight or branched hexyl) monosulfide and di(straight or branched hexyl) disulfide; dicyclohexyl sulfides such as dicyclohexyl monosulfide and dicyclohexyl disulfide; di(straight or branched heptyl) sulfides such as di(straight or branched heptyl) monosulfide and di(straight or branched heptyl) disulfide; di-tert-octyl sulfides such as di-tert-octyl monosulfide and di-tert-octyl disulfide; di(straight or branched octyl) sulfides such as di(straight or branched octyl) monosulfide and di(straight or branched octyl) disulfide; di(straight or branched nonyl) sulfides such as di(straight or branched nonyl) monosulfide and di(straight or branched nonyl) disulfide; di(straight or branched decyl) sulfides such as di(straight or branched decyl) monosulfide and di(straight or branched decyl) disulfide; di(straight or branched undecyl) sulfides such as di(straight or branched undecyl) monosulfide and di(straight or branched undecyl) disulfide; di-tert-dodecyl sulfides such as di-tert-dodecyl monosulfide and di-tert-dodecyl disulfide; di(straight or branched dodecyl) sulfides such as di(straight or branched dodecyl) monosulfide and di(straight or branched dodecyl) disulfide; di(straight or branched tridecyl) sulfides such as di(straight or branched tridecyl) monosulfide and di(straight or branched tridecyl) disulfide; di(straight or branched tetradecyl) sulfides such as di(straight or branched tetradecyl) monosulfide and di(straight or branched tetradecyl) disulfide; di(straight or branched pentadecyl) sulfides such as di(straight or branched pentadecyl) monosulfide and di(straight or branched pentadecyl) disulfide; di-2-methylpentadecyl sulfides such as di-2-methylpentadecyl monosulfide and di-2-methylpentadecyl disulfide; di-tert-hexadecyl sulfides such as di-tert-hexadecyl monosulfide and di-tert-hexadecyl disulfide; di(straight or branched hexadecyl) sulfides such as di(straight or branched hexadecyl) monosulfide and di(straight or branched hexadecyl) disulfide; di(straight or branched heptadecyl) sulfides such as di(straight or branched heptadecyl) monosulfide and di(straight or branched heptadecyl) disulfide; di(straight or branched octadecyl) sulfides such as di(straight or branched octadecyl) monosulfide and di(straight or branched octadecyl) disulfide; di(straight or branched nonadecyl) sulfides such as di(straight or branched nonadecyl) monosulfide and di(straight or branched nonadecyl) disulfide; di(straight or branched icosyl) sulfides such as di(straight or branched icosyl) monosulfide and di(straight or branched icosyl) disulfide; di(straight or branched heniceryl) sulfides such as di(straight or branched heniceryl) monosulfide and di(straight or branched heniceryl) disulfide; di(straight or branched docosyl) sulfides such as di(straight or branched docosyl) monosulfide and di(straight or branched docosyl) disulfide; dipropenyl sulfides such as dipropenyl monosulfide and dipropenyl disulfide; dibutenyl sulfides such as dibutenyl monosulfide and dibutenyl disulfide; diphenyl sulfides such as diphenyl monosulfide and diphenyl disulfide; dinaphthyl sulfides such as dinaphthyl monosulfide and dinaphthyl disulfide; ditolyl sulfides such as ditolyl monosulfide and ditolyl disulfide; diethylphenyl sulfides such as diethylphenyl monosulfide and diethylphenyl disulfide; dipropylphenyl sulfides such as dipropylphenyl monosulfide and dipropylphenyl disulfide; dibutylphenyl sulfides such as dibutylphenyl monosulfide and dibutylphenyl disulfide; dipentylphenyl sulfides such as dipentylphenyl monosulfide and dipentylphenyl disulfide; dihexylphenyl sulfides such as dihexylphenyl monosulfide and dihexylphenyl disulfide; diheptylphenyl sulfides such as diheptylphenyl monosulfide and diheptylphenyl disulfide; dioctylphenyl sulfides such as dioctylphenyl monosulfide and dioctylphenyl disulfide; dinonylphenyl sulfides such as dinonylphenyl monosulfide and dinonylphenyl disulfide; didodecylphenyl sulfides such as didodecylphenyl monosulfide and didodecylphenyl disulfide; diundecylphenyl sulfides such as diundecylphenyl monosulfide and diundecylphenyl disulfide; didodecylphenyl sulfides such as didodecylphenyl monosulfide and didodecylphenyl disulfide; dixylyl sulfides such as dixylyl monosulfide and dixylyl disulfide; diethylmethylphenyl sulfides such as diethylmethylphenyl monosulfide and diethylmethylphenyl disulfide; di(diethylphenyl) sulfides such as di(diethylphenyl) monosulfide and di(diethylphenyl) disulfide; di(dipropylphenyl) sulfides such as di(dipropylphenyl) monosulfide and di(dipropylphenyl) disulfide; di(dibutylphenyl) sulfides such as di(dibutylphenyl) monosulfide and di(dibutylphenyl) disulfide; dimethylnaphthyl sulfides such as dimethylnaphthyl monosulfide and dimethylnaphthyl disulfide; diethylnaphthyl sulfides such as diethylnaphthyl monosulfide and diethylnaphthyl disulfide; dipropylnaphthyl sulfides such as dipropylnaphthyl monosulfide and dipropylnaphthyl disulfide; dibutylnaphthyl sulfides such as dibutylnaphthyl monosulfide and dibutylnaphthyl disulfide; dibenzyl sulfides such as dibenzyl monosulfide and dibenzyl disulfide; diphenethyl sulfides such as diphenethyl monosulfide and diphenethyl disulfide; diphenylpropyl sulfides such as diphenylpropyl monosulfide and diphenylpropyl disulfide and a

mixture thereof.

[0096] Further another examples of the sulfides are diester sulfides represented by the formula

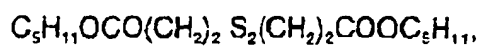
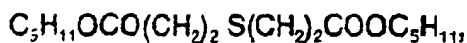
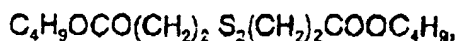


(XV)

[0097] In formula (XV), R^{32} and R^{33} are independently a straight or branched alkyl group having 2 to 20 carbon atoms, preferably 4 to 20 carbon atoms; c is a numeral of 1 or 2, preferably 1; and d and e each are a numeral of 2 to 5, preferably 2.

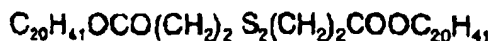
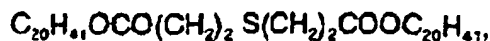
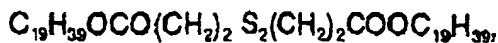
[0098] Examples of R^{32} and R^{33} include independently ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl straight or branched pentyl, straight or branched hexyl, straight or branched heptyl, straight or branched octyl, straight or branched nonyl, straight or branched decyl, straight or branched undecyl, straight or branched dodecyl, straight or branched tridecyl, straight or branched tetradecyl, straight or branched pentadecyl, straight or branched hexadecyl, straight or branched heptadecyl, straight or branched octadecyl, straight or branched nonadecyl and straight or branched icosyl.

[0099] Preferred examples of the diester sulfides include the compounds represented by the following formulae and a mixture of these compounds:

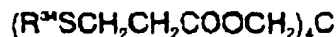


	$C_6H_{13}OCO(CH_2)_2 S(CH_2)_2 COOC_6H_{13},$
5	$C_6H_{13}OCO(CH_2)_2 S_2(CH_2)_2 COOC_6H_{13},$
	$C_7H_{15}OCO(CH_2)_2 S(CH_2)_2 COOC_7H_{15},$
	$C_7H_{15}OCO(CH_2)_2 S_2(CH_2)_2 COOC_7H_{15},$
10	$C_8H_{17}OCO(CH_2)_2 S(CH_2)_2 COOC_8H_{17},$
	$C_8H_{17}OCO(CH_2)_2 S_2(CH_2)_2 COOC_8H_{17},$
	$C_9H_{19}OCO(CH_2)_2 S(CH_2)_2 COOC_9H_{19},$
15	$C_9H_{19}OCO(CH_2)_2 S_2(CH_2)_2 COOC_9H_{19},$
	$C_{10}H_{21}OCO(CH_2)_2 S(CH_2)_2 COOC_{10}H_{21},$
20	$C_{10}H_{21}OCO(CH_2)_2 S_2(CH_2)_2 COOC_{10}H_{21},$
	$C_{11}H_{23}OCO(CH_2)_2 S(CH_2)_2 COOC_{11}H_{23},$
	$C_{11}H_{23}OCO(CH_2)_2 S_2(CH_2)_2 COOC_{11}H_{23},$
25	$C_{12}H_{25}OCO(CH_2)_2 S(CH_2)_2 COOC_{12}H_{25},$
	$C_{12}H_{25}OCO(CH_2)_2 S_2(CH_2)_2 COOC_{12}H_{25},$
	$C_{13}H_{27}OCO(CH_2)_2 S(CH_2)_2 COOC_{13}H_{27},$
30	$C_{13}H_{27}OCO(CH_2)_2 S_2(CH_2)_2 COOC_{13}H_{27},$
	$C_{14}H_{29}OCO(CH_2)_2 S(CH_2)_2 COOC_{14}H_{29},$
35	$C_{14}H_{29}OCO(CH_2)_2 S_2(CH_2)_2 COOC_{14}H_{29},$
	$C_{15}H_{31}OCO(CH_2)_2 S(CH_2)_2 COOC_{15}H_{31},$
	$C_{15}H_{31}OCO(CH_2)_2 S_2(CH_2)_2 COOC_{15}H_{31},$
40	$C_{16}H_{33}OCO(CH_2)_2 S(CH_2)_2 COOC_{16}H_{33},$
	$C_{16}H_{33}OCO(CH_2)_2 S_2(CH_2)_2 COOC_{16}H_{33},$
45	$C_{17}H_{35}OCO(CH_2)_2 S(CH_2)_2 COOC_{17}H_{35},$
	$C_{17}H_{35}OCO(CH_2)_2 S_2(CH_2)_2 COOC_{17}H_{35},$
	$C_{18}H_{37}OCO(CH_2)_2 S(CH_2)_2 COOC_{18}H_{37},$
50	$C_{18}H_{37}OCO(CH_2)_2 S_2(CH_2)_2 COOC_{18}H_{37},$
	$C_{19}H_{39}OCO(CH_2)_2 S(CH_2)_2 COOC_{19}H_{39},$

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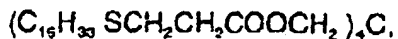
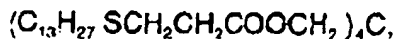
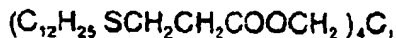
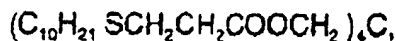
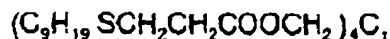
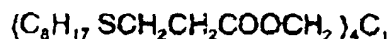
[0100] Still another examples of the sulfides are tetraester tetrasulfides represented by the formula

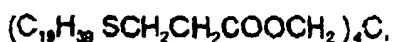
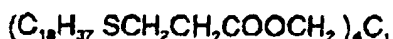


(XVI)

[0101] In formula (XVI), R^{34} is a straight or branched alkyl group having 8 to 20 carbon atoms or preferably 10 to 18 carbon atoms. Examples of R^{34} include straight or branched octyl, straight or branched nonyl, straight or branched decyl, straight or branched undecyl, straight or branched dodecyl, straight or branched tridecyl, straight or branched tetradecyl, straight or branched pentadecyl, straight or branched hexadecyl, straight or branched heptadecyl, straight or branched octadecyl, straight or branched nonadecyl and straight or branched icosyl. Among these groups, straight or branched dodecyl and straight or branched tridecyl are preferred.

[0102] Preferred examples of the tetraester tetrasulfides include compounds represented by the following formulae and a mixture of these compounds:





[0103] Among these compounds, the compound represented by $(C_{12}H_{25}SCH_2CH_2COOCH_2)_4C$ is particularly preferred.

15 [0104] Needless to mention, Component (B) which serves as an oxidation inhibitor in the inventive lubricating oil composition may be a single compound selected from the aforementioned hindered phenol-based, aromatic amine-based and sulfur containing compounds, or alternatively, may be a mixture of two or more of the compounds at any ratio therebetween.

[0105] However, Component (B) is preferably a single compound or a mixture of two or more compounds at any ratio
20 therebetween selected from the above-described hindered phenol compounds, aromatic amine compounds and metal-containing sulfur containing compounds such as metal salts of dihydrocarbyldithiophosphoric acid and metal salts of dithiocarbamic acid when consideration given to a high synergistic effect on oxidation stability achieved by the combination of Component (A) and Component (B). Meanwhile, in the case where a metal-free sulfur-based oxidation inhibitor is used, it is preferably used in combination with a single compound or a mixture of two or more compounds selected
25 from the above-described hindered phenol compounds, aromatic amine compounds and metal and sulfur-containing compounds such as metal salts of dihydrocarbyldithiophosphoric acid and metal salts of dithiocarbamic acid.

[0106] Although the content at the component (B) in the lubricating oil composition of the present invention may be selected at will ordinarily, the lower limit is 0.1 % by mass, preferably 0.2% by mass, based on the total amount of the lubricating oil composition, while the upper limit is 5.0% by mass, preferably 2.0% by mass, based on the total amount
30 of the lubricating oil composition.

[0107] Contents less than 0.1% by mass would fail in improved oxidation stability, while contents more than 5.0% would be economically infeasible with no particular improvement in oxidation stability.

[0108] Although the lubricating oil composition of the present invention per se is excellent in oxidation stability, in order to further enhance the performances of the lubricating oil composition of the present invention, it may be blended with
35 one or more suitable additives. Eligible additives includes detergents such as a neutral, basic, calcium carbonate per-basic, or calcium borate perbasic alkaline earth metal sulfonate, an alkaline earth metal salicylate and an alkaline earth metal phenolate; ashless dispersants such as polybutenyl succinimide, a polyamine having a long alkyl chain, an amide formed from a higher fatty acid and a polyamine and borides thereof; wear inhibitors such as organic phosphate, organic phosphorus ester, a fatty acid, an ester of fatty acid and an aliphatic alcohol; friction reducing agents such as molybdenum dithiophosphate, molybdenum disulfide, a higher aliphatic amine, a higher fatty acid, an ester of higher
40 fatty acid and a higher aliphatic alcohol; a rust inhibitor such as petroleum sulfonate, alkylbenzene sulfonate, dinonylnaphthalene sulfonate, an ester of alkenyl succinate and an ester of polyhydric alcohol; a demulsifier exemplified by a polyalkylene glycol-based nonionic surfactant such as polyoxyethylene alkyl ether, polyoxyethylene alkylphenyl ether and polyoxyethylene alkyl-naphthyl ether; metal deactivator such as imidazoline, a pyrimidine derivative, alkylthiazole, mercaptobenzothiazole, benzotriazole or a derivative thereof, 1,3,4-thiadiazole polysulfide, 1,3,4-thiadiazolyl-2,5-bisdi-alkyl dithiocarbamate, 2-(alkyldithio)benzoimidazole and β -(o-carboxybenzylthio)propionitrile; antifoamers such as silicone, fluorosilicone and fluoroalkyl ether; viscosity index improvers such as polymethacrylate, an olefinic copolymer and a hydrogenation product thereof; and pour point depressants such as polymethacrylate.

[0109] In the case where the above-described additives are added to the lubricating oil composition of the present invention, the amount of each additive is selected arbitrary. Preferred examples of such amounts are as follows: the
50 amount of the antifoamers is in the range of from 0.0005 to 1 part by mass, the amount of the metal deactivator is in the range of from 0.005 to 1 part by mass, and the amount of each of the other agent is in the range of from 0.1 to 15 parts by mass, based on 100 parts by mass of the lubricating oil composition.

[0110] The lubricating oil composition according to the invention finds wide range of applications such as lubricating
55 oils for internal combustion engines such as gasoline engines of a two-wheeled vehicle and a four-wheeled vehicle and diesel engines for land use or marine use; automotive gear oils for automatic or manual transmission and differential; gear oils for industrial use; turbine oils; hydraulic oils; oils for compressors and refrigerators; oils for plastic processing such as cutting, rolling, pressing, forging, squeezing, drawing and punching; oils for metal processing such as heating

and electric discharging; oils for guide sliding surface; bearing oils; rust preventing oils and heat transfer oils.

EXAMPLES

- 5 [0111] In order to better explain the present invention, the following examples and comparative examples are given by way of illustration and not by way of limitation.

Examples 1 to 7 and Comparative Examples 1 to 5

- 10 [0112] Various lubricating oil compositions listed as Examples 1 - 7 and Comparative Examples 1 - 5 in Tables 1 and 2 were prepared from the following formulations. The oxidation stability test was conducted in accordance with Rotary Bomb Oxidation Test stipulated by ASTM D-2272 to measure time length from initiation of oxidation to termination thereof. The results are shown in the bottom columns of Tables.

[0113] The lubricating base oils and the oxidation inhibitors are identified as follows:

15

Component (A): mineral oil-derived lubricating base oils

Refined mineral oil A

20 [0114]

saturated hydrocarbon component : 96.1% by mass
viscosity-density constant of saturated hydrocarbon component : 0.781
kinetic viscosity at 100°C : 4.155 mm² /s
25 kinetic viscosity at 40°C : 18.73 mm² /s
viscosity index : 126
fluid point : -22.5°C

Refined mineral oil B

30

[0115]

saturated hydrocarbon component : 93.9% by mass
viscosity-density constant of saturated hydrocarbon component : 0.772
35 kinetic viscosity at 100°C : 4.337 mm² /s
kinetic viscosity at 40°C : 19.88 mm² /s
viscosity index : 128
fluid point : -22.5°C

40 Refined mineral oil C

[0116]

saturated hydrocarbon component : 73.2% by mass
viscosity-density constant of saturated hydrocarbon component : 0.781
45 kinetic viscosity at 100°C : 4.394 mm² /s
kinetic viscosity at 40°C : 21.65 mm² /s
viscosity index : 112
fluid point : -15°C

50

Refined mineral oil D

[0117]

saturated hydrocarbon component : 98.9% by mass
viscosity-density constant of saturated hydrocarbon component : 0.806
55 kinetic viscosity at 100°C : 4.176 mm² /s
kinetic viscosity at 40°C : 20.59 mm² /s

viscosity index : 105

fluid point : -12.5°C

Component (B): oxidation inhibitors

[0118]

Hindered phenol type A : 2,6-di-tert-butyl-p-cresol

Hindered phenol type B : 2,6-di-tert-butyl-4-ethylphenol

Aromatic amine type A : phenyl- α -naphthylamine

Aromatic amine type B : p,p'-di(branched nonyl)diphenylamine

Sulfur containing compound type A : zinc dialkyldithiophosphate (alkyl being a mixture of sec-butyl and sec-hexyl)

Sulfur containing compound type B : dibenzyl disulfide

Sulfur containing compound type C : dilauryl thiodipropionate

[0119] It can be seen from the results of Table 1 that all of the lubricating oil compositions (Examples 1 to 7) of the present invention exhibit excellent oxidation stability.

[0120] To the contrary, markedly poor oxidation stability was observed in Comparative Example 1 in which the lubricating oil composition was composed solely of the base oil of the present invention without the addition of any oxidation inhibitor; in Comparative Example 2 in which the lubricating oil composition was composed solely of the refined mineral oil C having a saturated hydrocarbon content of less than 80% by mass in place of the base oil of the present invention; and in Comparative Example 4 in which the lubricating oil composition was composed solely of the refined mineral oil D whose saturated hydrocarbon component had a viscosity-density constant of more than 0.79.

[0121] Besides, as is apparent from Comparative Examples 3 and 5, even if an oxidation inhibitor is added to an oil which is not the base oil of the present invention but the refined mineral oil C having a saturated hydrocarbon content of less than 80% by mass or to an oil which is not the base oil at the present invention but is the refined mineral oil D having a viscosity-density constant of saturated hydrocarbon of more than 0.79, the oxidation stability of these oils cannot be remarkably improved. In short, the oxidation stability of a lubricating oil composition can be improved if the saturated hydrocarbon content of the base oil of the lubricating oil composition is within a specific range and the viscosity-density constant of the saturated hydrocarbon component is within a specific range. That is, if Example 1, Comparative Examples 3 and 5, and Comparative Examples 1, 2, and 4 are compared with one another, it becomes clear that oxidation stability is remarkably improved only in the case where an oxidation inhibitor is added to the base oil of the present invention but otherwise oxidation stability is not improved. Accordingly, in the case where an oxidation inhibitor is added to a base oil of the present invention, the improvement of the oxidation stability is clearly based on the synergism of the base oil and the oxidation inhibitor.

[0122] As stated above, it is possible to obtain a lubricating oil composition by specifying the saturated hydrocarbon content of base oil and the viscosity-density constant of the saturated hydrocarbon component.

Table 1

	Example 1	Example 2	Example 2	Example 3	Example 4	Example 5	Example 6	Example 7
Composition of base oil, % by mass	99.4	99.0	99.5	99.0	99.0	99.3	99.3	99.4
Oxidation inhibitor, % by mass	0.6	1.0	0.5	1.0	1.0	0.6	0.6	0.6
Oxidative stability test (min.) ^{*)}	390	380	5000	2800	600	530	470	303

^{*)} conducted in accordance with JIS K2514 3.3 to measure time length from initiation to termination.

Table 2

	Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4	Comparative Example 5
Composition of base oil, % by mass	100	100	99.4	100	99.4
Oxidation inhibitor, % by mass			0.6		0.6
Oxidative stability test (min.) ^{*)}	<30	<30	160	<30	244

^{*)} conducted in accordance with ASTM D-2272 to measure time length from initiation to termination.

Claims

1. A lubricating oil composition comprising a base oil and an oxidation inhibitor, wherein the lubricating base oil is a mineral oil-derived lubricating oil having a saturated hydrocarbon content of 80% by mass or more and the viscosity-density constant of the saturated hydrocarbon component is 0.79 or less.

2. A lubricating oil composition according to claim 1 wherein said lubricating base oil has a kinetic viscosity at 40°C in the range of 5 mm²/s to 200 mm²/s.

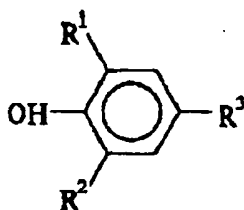
3. A lubricating oil composition according to claim 1 wherein said lubricating base oil has a kinetic viscosity at 100°C in the range of 0.5 mm²/s to 100 mm²/s.

4. A lubricating oil composition according to claim 1 wherein said lubricating base oil is 50 or more in viscosity index.

5. A lubricating oil composition according to claim 1 wherein said lubricating base oil is 0°C or lower in pour point.

6. A lubricating oil composition according to claim 1 wherein said oxidation inhibitor is at least one member selected from the group consisting of a hindered phenol compound, an aromatic amine compound and a sulfur containing compound.

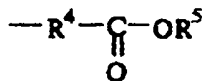
7. A lubricating oil composition according to claim 6 wherein said hindered phenol compound is one or more members selected from the group consisting of compounds represented by the formula



(I)

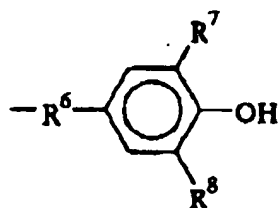
wherein R¹ and R² each are an alkyl group having 1 to 4 carbon atoms with the proviso that at least one of R¹ and R² is a tert-butyl group, and R³ is selected from the group consisting of an alkyl group having 1 to 4 carbon atoms and groups represented by formulae (II), (III) and (IV);

said formula (II) being represented by

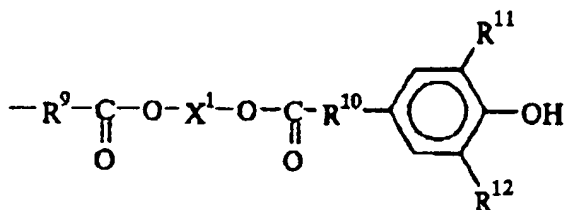


wherein R⁴ is an alkylene group having 1 to 6 carbon atoms and R⁵ is an alkylene group having 1 to 24 carbon groups;

said formula (III) being represented by



wherein R^6 is an alkylene group having 1 to 6 carbon atoms and R^7 and R^8 each are an alkyl group having 1 to 4 carbon atoms with the proviso that at least one of R^7 and R^8 is a tert-butyl group; and said formula (IV) being represented by



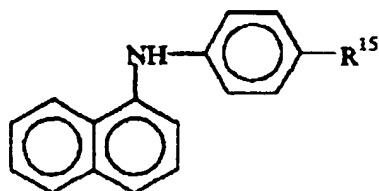
wherein R^9 and R^{10} each are an alkylene group having 1 to 4 carbon atoms, R^{11} and R^{12} each are an alkyl group having 1 to 4 carbon atoms with the proviso that at least one of R^{11} and R^{12} is a tert-butyl group and X^1 is selected from the group consisting of an alkylene group having 1 to 18 carbon atoms and a group represented by the formula



(V)

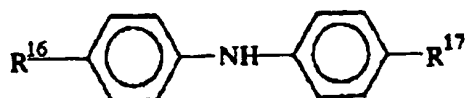
wherein R^{13} and R^{14} each are an alkylene group having 1 to 6 carbon atoms.

7. A lubricating oil composition according to claim 6 wherein said aromatic amine compound is one or more members selected from phenyl- α -naphthylamine, N-alkylphenyl- α -naphthylamine represented by the formula



(VII)

wherein R^{15} is a straight or branched alkyl group having 1 to 16 carbon atoms; and p, p'-dialkyldiphenylamine represented by the formula

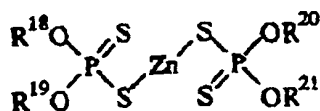


(IX)

wherein R^{16} and R^{17} each are a straight or branched alkyl group having 1 to 16 carbon atoms.

8. A lubricating oil composition according to claim 6 wherein said sulfur containing compound is one or more member selected from metal salts of dihydrocarbyldithiophosphoric acid, metal salts of dithiocarbamic acid and sulfides.

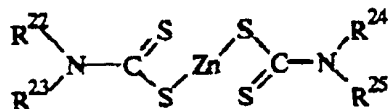
9. A lubricating oil composition according to claim 8 wherein said metal salts of dihydrocarbyldithiophosphoric acid are one member selected from the group consisting of zinc dihydrocarbyldithiophosphates represented by the formula



(X)

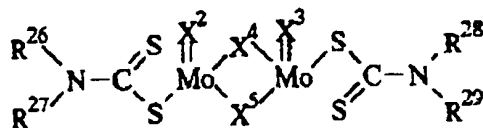
wherein R^{18} , R^{19} , R^{20} and R^{21} each are a hydrocarbon group having 1 to 18 carbon atoms.

10. A lubricating oil composition according to claim 8 wherein said metal salts of dithiocarbamic acid are one member selected from the group consisting of zinc dihydrocarbyldithiocarbamates represented by the formula



(XI)

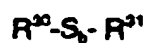
wherein R^{22} , R^{23} , R^{24} and R^{25} each are a hydrocarbon group having 1 to 18 carbon atoms; and molybdenum dihydrocarbyldithiocarbamates represented by the formula



(XII)

wherein R^{26} , R^{27} , R^{28} and R^{29} each are a hydrocarbon group having 1 to 18 carbon atoms and X^2 , X^3 , X^4 and X^5 each are selected from the group consisting of a sulfur atom and an oxygen atom.

10. A lubricating oil composition according to claim 8 wherein said sulfides are one member selected from the group consisting of dihydrocarbyl sulfides represented by the formula



(XIV)

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wherein R^{30} and R^{31} each are selected from a straight or branched alkyl group having 1 to 22 carbon atoms, an aryl group having 6 to 20 carbon atoms, an alkylaryl group having 7 to 20 carbon atoms and an arylalkyl group having 7 to 20 carbon atoms and b is a numeral of 1 to 5; diester sulfides represented by the formula

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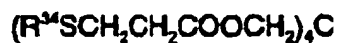


(XV)

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wherein R^{32} and R^{33} each are a straight or branched alkyl group having 2 to 20 carbon atoms, c is a numeral of 1 or 2 and d and e each are a numeral of 2 to 5; and tetraester tetrasulfides represented by the formula

20



(XVI)

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wherein R^{34} is a straight or branched alkyl group having 8 to 20 carbon atoms.

11. A lubricating oil composition according to claim 8 wherein the content of said oxidation inhibitor is in the range of 0.1 to 2.0 % by mass based on the total amount of the said lubricating oil composition.

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European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 98 85 0129

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	EP 0 281 992 A (IDEMITSU KOSAN CO) 14 September 1988 * page 3, line 3 - page 4, line 20 * ---	1-7,9,11	C10M101/02 C10M169/04 //(C10M169/04, 101:02,129:10, 129:14,129:76, 133:12,135:18, 135:22,135:24, 135:26,135:28, 137:10), C10N10:04, C10N10:12, C10N30:10, C10N40:00
X	WO 95 27022 A (EXXON RESEARCH ENGINEERING CO ;TOMIZAWA HIROTAKA (JP); ARAI KATSUY) 12 October 1995 * page 2, paragraph 4 - page 4, paragraph 1 * ---	1-8,10,11	
X	EP 0 435 670 A (NIPPON OIL CO LTD) 3 July 1991 * page 4, line 8 - page 6, line 13 * * page 7, line 9 - line 21 * ---	1-9,11	
X	GB 1 413 677 A (SUN OIL CO PENNSYLVANIA) 12 November 1975 * page 1, line 46 - line 75 * ---	1-7,9,11	
X	EP 0 725 130 A (TONEN CORP) 7 August 1996 * page 2, line 46 - page 4, column 34 * ---	1-11	TECHNICAL FIELDS SEARCHED (Int.Cl.6)
A	GB 2 083 494 A (SHELL INT RESEARCH) 24 March 1982 * page 2, line 19 - page 3, line 38 * -----	1-5	C10M
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 23 November 1998	Examiner Rotsaert, L
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>			

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